# Evaluation of the Anisotropy Field for Fine-Particle Systems from Low-Field Thermomagnetic Curves

J. Geshev, Ll. M. Martinez, J. S. Muñoz, J. E. Schmidt, and M. Mikhov

***Abstract*— A method for evaluating the temperature-dependent magnetic anisotropy field from thermomagnetic curves for Stoner–Wohlfarth-like systems is proposed. It allows the anisotropy parameters of a sample with unknown spontaneous magnetization to be obtained by using magnetic fields considerably lower than the anisotropy field.**

***Index Terms*— Fine-particle systems, magnetic anisotropy, single-domain particles, thermomagnetic curves.**

## I. Introduction

THE magnetic anisotropy is one of the most important characteristics of fine-particle materials. It determines the magnetic state of the particle systems in different external conditions, temperature and magnetic fields, and their applicability for a number of practical uses such as permanent magnets, magnetic recording media, etc. The investigation of magnetic anisotropy is one of the main problems of experimental magnetism and a variety of methods exist for this purpose [1]–[14]. Among them are: torque curves measurements and rotational hysteresis loss [3]–[5], singular-point detection [6], reversible transverse susceptibility [7], [8], ferromagnetic resonance [9], torsion pendulum [10]–[12], demagnetization curves (DMC) fit [13], and remanence curves [14].

Unfortunately, to evaluate the anisotropy field $H_a$ of a disordered fine-particle system, all these methods require the use of fields exceeding $H_a$. The anisotropy fields of modern hard magnetic materials are of the order of several hundreds of kOe, and such fields are only available in a limited number of laboratories in the world.

As shown in [15]–[20], low-field thermomagnetic curves (i.e., magnetization versus temperature dependence at a fixed low magnetic field) for disordered systems of single-domain particles can exhibit Hopkinson-type behavior, due to the specific temperature dependencies of both the spontaneous magnetization $M_s(T)$ and the magnetic anisotropy constant $K_1(T)$. In fact, the Hopkinson-type effect (existence of a peak in the initial thermomagnetic curves near the Curie temperature $T_c$) observed in many different systems and related to different mechanisms, is always governed by the temperature dependence of magnetic anisotropy. It means that the low-field thermomagnetic curves contain information on the temperature dependencies of both the spontaneous magnetization and the magnetic anisotropy.

In the present work, a method for estimation of the temperature dependence of the anisotropy field from low-field thermomagnetic curves is proposed for Stoner–Wohlfarth (S–W) [21] systems. Many real particulate assemblies can be considered to a certain extent as S–W-like ones.

The main advantage of the method is that it allows the temperature dependence of the anisotropy field of a sample with unknown spontaneous magnetization to be obtained, using magnetic fields lower than $H_a$.

## II. Physical Background of the Method

Consider a S–W system [21] consisting of noninteracting, single-domain, randomly orientated uniaxial particles. The magnetic state of the system can change by coherent rotation of the particles' magnetic moments only. All particles have the same volume $V$ and shape. $N_{||}$ and $N_{\perp}$ denote the demagnetization factors of the particles along the axes directed parallel and perpendicular to their crystallographic easy magnetization axis $c$, respectively. The spontaneous magnetization of the particles is $M_s$ and their magnetocrystalline anisotropy is described by the first two anisotropy constants $K_1$ and $K_2$. The effective first order anisotropy constant of the particles $K_u$ is

$$K_u = K_1 - \frac{1}{2}(N_{||} - N_{\perp})M_s^2. \tag{1}$$

The reduced free energy of a particle $\eta = E/(2K_u + 4K_2)$, placed in magnetic field **H**, neglecting thermal activation effects, can be written as

$$\eta = \frac{1}{2 + 4K_2/K_u}\left(\sin^2\theta + \frac{K_2}{K_u}\sin^4\theta\right) - \frac{H}{H_a}\cos(\psi - \theta) \tag{2}$$

where $\theta$ is the angle between $\mathbf{M}_s$ and $c$, $\psi$ the angle between **H** and $c$, and the anisotropy field equals

$$H_a = (2K_u + 4K_2)/M_s. \tag{3}$$

The equilibrium direction of the magnetization vector can be calculated by minimizing $\eta$ for a given parameter set

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$(K_2/K_u, H_a, H, \psi)$. The reduced magnetization $j = M/M_s$ of an assembly of randomly orientated or aligned particles is given by the weighted average of the projections of the magnetization vectors of the individual particles along the external field direction, so the initial magnetization curve (IMC) and the hysteresis loop for such a system can be calculated. When $M_s(T)$, $K_u(T)$, and $K_2(T)$ are known, the model zero-field-cooled (ZFC) and field-cooled (FC) thermomagnetic curves for a S–W-like system can be obtained as well. Thus, it is a possible to obtain the anisotropy parameters by fitting the experimentally measured magnetization to the calculated values.

Magnetization curves have already been used for estimation of magnetic anisotropy parameters for disordered and textured particulate samples by using fitting procedures [13], [22], [23]. The values of the anisotropy constants $K_1$ and $K_2$ are varied until the calculated demagnetization curve (DMC) is fitted to the experimental one. The saturation magnetization of the sample is considered as a known parameter, or it is varied as a third one.

Combinations of IMC and DMC, as well as ZFC and FC thermomagnetic curves, have not yet been used for estimation of magnetic anisotropy parameters.

For a S–W system, when the anisotropy field decreases monotonically with temperature (which is normally the case), there exists a direct correspondence between ZFC and FC thermomagnetic curves on one hand and the initial magnetization curve and the demagnetizing branch of an hysteresis loop on the other (this relation is explained in the Appendix). At a given temperature, each pair of points on the ZFC and the FC thermomagnetic curves, plotted in a given magnetic field, corresponds to a pair of points on the IMC and the DMC for the same field, plotted at the same temperature. This is demonstrated in Fig. 1 for model thermomagnetic and magnetization curves, calculated for M-type Ba-ferrite single-domain particles. There exists a direct correspondence between the ratio $R(H) = M^{\mathrm{IMC}}(H)/M^{\mathrm{DMC}}(H)$ (calculated for a couple of magnetization curves, plotted at a given temperature) and the ratio $R(T) = M^{\mathrm{ZFC}}(T)/M^{\mathrm{FC}}(T)$ (taken for the same temperature from the thermomagnetic curves, plotted in the same magnetic field). Obviously, $R$ is independent of the saturation magnetization. The temperature and field dependencies of the above ratios, i.e., $R(T/T_c)$ and $R(H/H_a)$, respectively, are shown in Fig. 1(a) and (b).

Usually, to estimate the anisotropy parameters by fitting a set of experimental thermomagnetic curves, three parameters, $M_s$, $K_u$, and $K_2$, must be varied for each temperature, as it is the case in the DMC method [13]. However, taking into account the relation between the (ZFC, FC) and (IMC, DMC) sets of curves mentioned above, one can reduce the number of the fitting parameters. In the method proposed in the present work, it is not necessary to know the spontaneous magnetization of the material, i.e., one parameter less is used in the fitting procedure. For this purpose, instead of fitting the demagnetization curves, the $R$ curves are fitted.

The reduced energy (2) depends on two parameters: $K_2/K_u$ and $H/H_a$. Fixing $K_2/K_u$, the reduced IMC and DCD can be calculated easily. A set of these curves, as well as

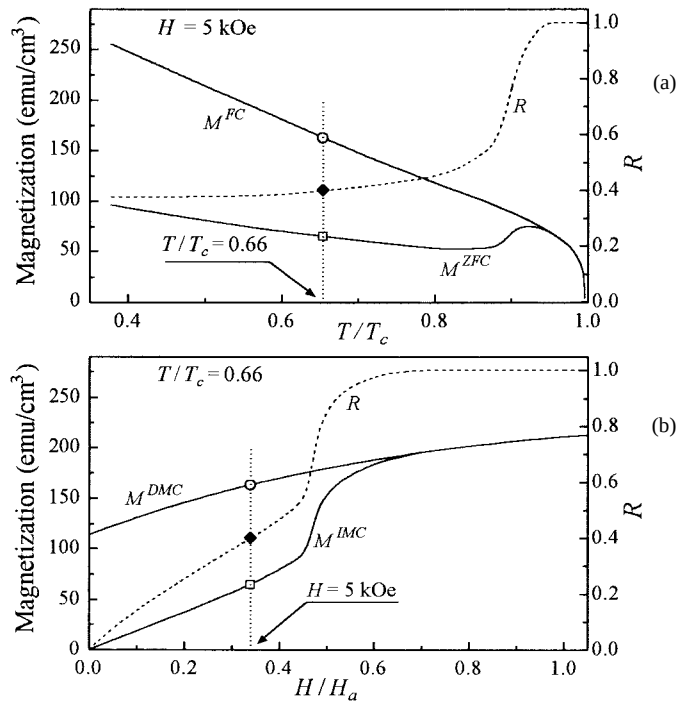


Fig. 1. (a) Model ZFC, FC, and the corresponding $R$ curves versus $T/T_c$ for $H = 5$ kOe. $M_s(T)$ and $K_u(T)$ are those for Ba-ferrite [25]. (b) Model IMC, DMC, and $R$ for the same system at $T/T_c = 0.66$.

the corresponding $R(H/H_a)$ curves are shown in Fig. 2(a) and (b), respectively, for different $K_2/K_u$. It can be seen that the shape of the curves changes continuously and they move downwards with the increase of $K_2/K_u$. Due to the monotonic change of the demagnetization curves, the shape of the $R(H/H_a)$ curves follows, to a large extent, the shape of the initial magnetization curves.

In order to check the method, it was applied on a sample of disordered Ba-ferrite fine particles that can be considered as a S–W system, for which the $H_a$ value is known [24]. The room temperature anisotropy field was determined by fitting the $R$ curve, $R(H) = M^{\mathrm{IMC}}(H)/M^{\mathrm{DMC}}(H)$, to the model one for $K_2 = 0$, as it is the case for Ba ferrite. The best fit was obtained for $H_a = 12.3$ kOe, in a very good agreement with the value of 12.0 kOe of the previous measurement [24]. The experimental $R(H/H_a)$ data are shown in Fig. 3, where the field was normalized by $H_a = 12.3$ kOe to fit the model curve for $K_2 = 0$.

The sequence of steps one should follow to estimate the temperature dependence of the anisotropy field using experimental low-field thermomagnetic curves is:

1) measurement of a set of ZFC and FC couples for different $H$;
2) calculation, for each temperature (for which the anisotropy field is to be estimated) the $R^{\mathrm{exp}}(H) = M^{\mathrm{ZFC}}/M^{\mathrm{FC}}$ for all $H$, i.e., from all thermomagnetic curve couples;
3) plotting this experimental $R^{\mathrm{exp}}$ versus $H/H_a$ dependence, varying $H_a$ until thus obtained $R^{\mathrm{exp}}(H/H_a)$ curve coincides reasonably well with one of the previously calculated $R^{\mathrm{calc}}(H/H_a)$ for different $K_2/K_u$

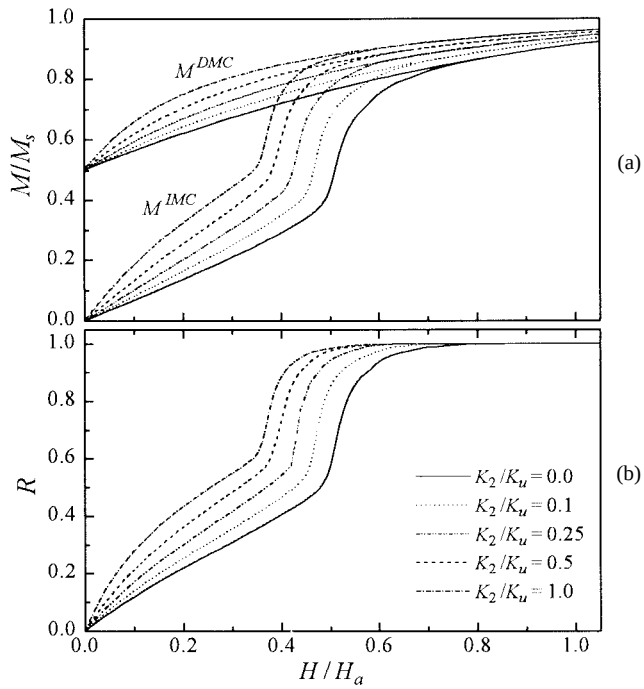


Fig. 2. (a) Model initial magnetization and demagnetization curves versus $H/H_a$ for a SW system for different $K_2/K_u$. (b) $R$ versus $H/H_a$ obtained from the curves in Fig. 2(a).

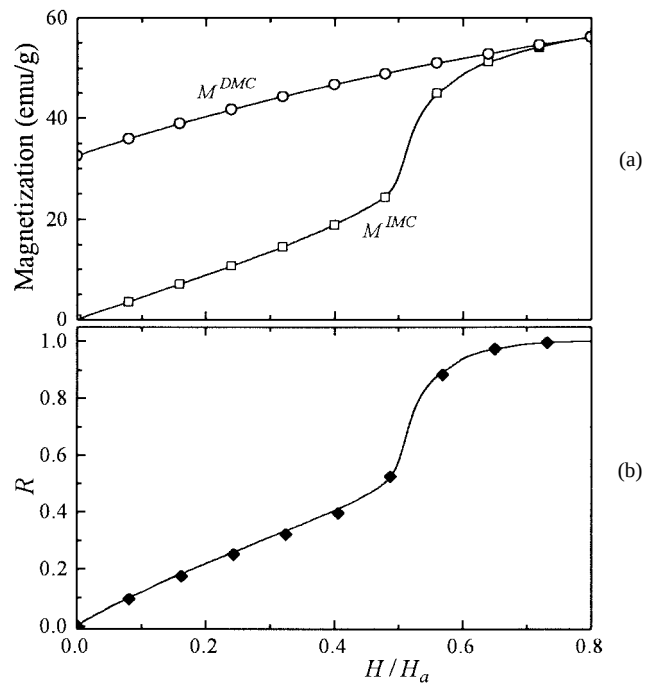


Fig. 3. (a) Experimental IMC and DMC versus $H/H_a$ for Ba-ferrite fine powder. (b) Model $R(H/H_a)$ curve for $K_2 = 0$ (solid line); the full symbols represent $R$ versus $H/H_a$ data obtained from the experimental curves in Fig. 3(a) for $H_a = 12.3$ kOe.

curves [Fig. 2(b)]. $H_a$ and $K_2/K_u$ obtained from the best fit are the anisotropy parameters.

A comparison of the accuracy of the DMC method and the present one is demonstrated in Fig. 4. The lines represent the model demagnetization curves [Fig. 4(a)] and the corresponding $R$ versus $H/H_a$ [Fig. 4(b)] for $K_2/K_u = 0.17$, $0.25$, $0.8$, and $1.0$. The circles and squares represent the modified data for the cases of $K_2/K_u = 0.25$ and $1.0$, as an error of 10% in $H_a$ is introduced ($H_a^{\text{modified}} = 1.1H_a$). It can be seen from the figure that even using a maximum field close to $H_a$, because of the similarity of the shape of the "neighbor" model DMC curves, the artificially erroneous data (circles and squares) fit reasonably well with the DMC curves for $K_2/K_u = 0.17$ and $0.8$, respectively [Fig. 4(a)]. However, the discrepancies between the erroneous $R$ data for $K_2/K_u = 0.25$ and $1.0$, and the model $R$-curves for $K_2/K_u = 0.17$ and $0.8$, respectively [Fig. 4(b)], become noticeable for rather lower fields (approximately half) than for the DMC method maximum fields.

The advantages of the present method are: 1) it allows the temperature dependence of the anisotropy field to be obtained using magnetic fields lower than $H_a$; 2) due to the specific shape of the $R$ curves, the number of the experimental points necessary for this method can be smaller than that used in the DMC method; and 3) it is not necessary to know the spontaneous magnetization of the material.

Obviously, the method cannot be applied in a temperature range where a magnetic phase transitions take place, as well as for temperatures at which ZFC and FC curves are very close. To reduce the effects of domain-wall magnetization processes for the larger particles in low fields, the use of very low-field magnetization data should be avoided, as in the DMC method. Nevertheless, the present method is useful because of its applicability to unknown samples.

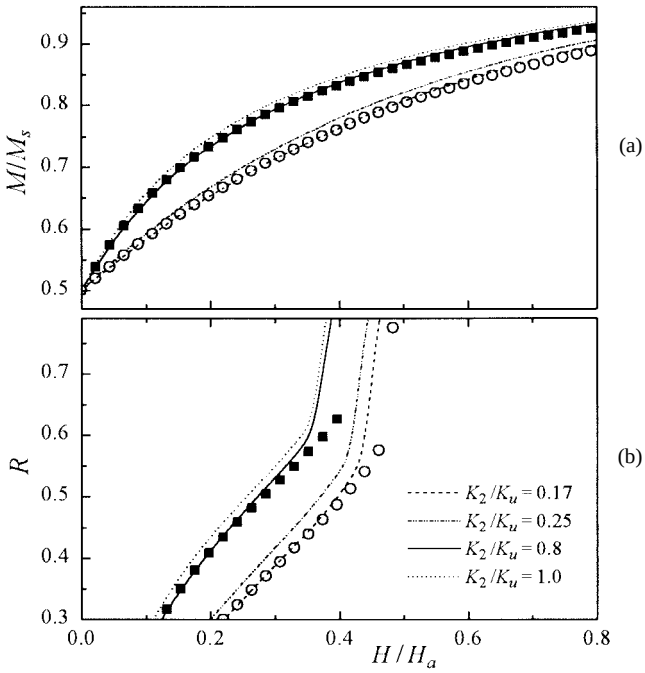


Fig. 4. (a) Model demagnetization curves for different $K_2/K_u$ and (b) the corresponding $R$ versus $H/H_a$. The squares and circles represent the modified data with $K_2/K_u = 1.0$ and $0.25$, as an error of 10% in $H_a$ is introduced ($H_a^{\text{modified}} = 1.1H_a$).

The method described above was applied for estimation of the temperature dependence of the anisotropy field of a real

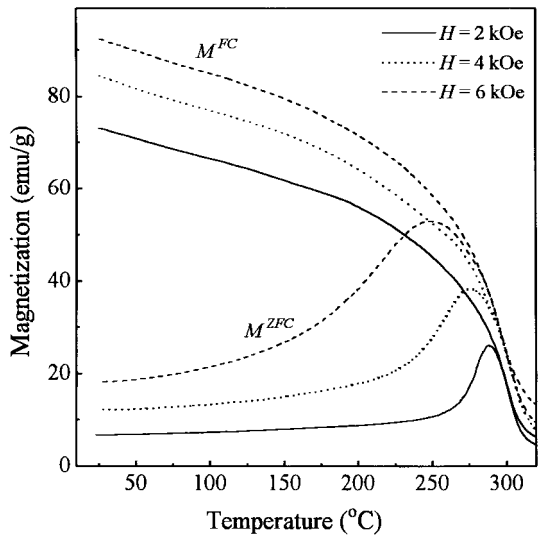


Fig. 5. Representative experimental FC and ZFC curves for a sample of melt-spun Nd–Fe–B ribbons (MQ powder).

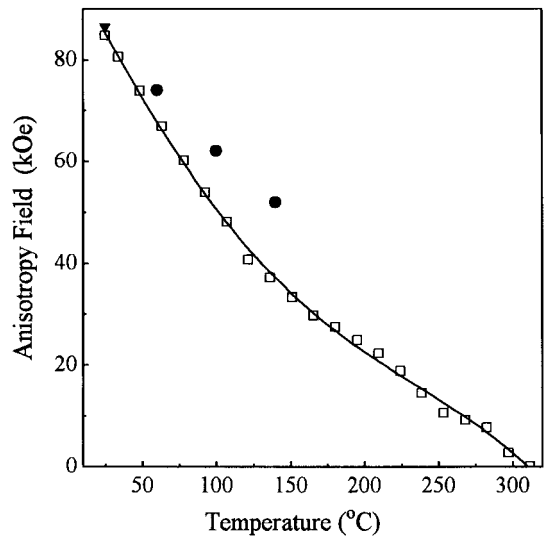


Fig. 6. Temperature dependence of the anisotropy field of the melt-spun Nd–Fe–B ribbons (MQ powder), obtained from the measured thermomagnetic curves shown in Fig. 5. The triangle represents the room temperature $H_a$ value obtained from [27]; the circles represent the $H_a$ values given in [23]. The line is a guide to the eye.

sample of hard magnetic material having fine grain structure. Commercially available pieces of melt-spun Nd–Fe–B ribbons, produced by General Motors and known as MQ powder, are used for this purpose since, according to [20], [26], and [27], they can be considered as consisting of single-domain particles.

Thermomagnetic curves were measured between room temperature and $T_c$ of the material in fields of 1, 2, 3, 4, 6, 8, and 11.5 kOe in vacuum using an Oxford Instruments vibrating sample magnetometer. Some of these thermomagnetic curves are shown in Fig. 5.

The temperature dependence of anisotropy field is presented in Fig. 6. For comparison, in the same figure are given the value of the anisotropy field calculated using the room temperature values of $K_u$, $K_2$, and $M_s$ from [27] and the values of $H_a(T)$ obtained from [23].

There are some differences between the anisotropy field estimated by the present method and the values found in the literature. These differences could be related to the fact that the literature data are for sintered Nd–Fe–B samples, whereas our measurements are on rapid-quenched ones. Another source of this discrepancy could be thermally activated changes of the particles' magnetization, which cannot be taken into account in the present work (as well as their shape anisotropy), since the particle size distribution is not available.

The present method can be applied, after slight modifications, for systems with different textures, as well as for systems with multiaxial magnetic particles.

## III. Conclusion

A new method for determination of the magnetic anisotropy of single-domain uniaxial particles is proposed. The temperature dependence of the anisotropy parameters is obtained from a set of ZFC and FC thermomagnetic curves, measured in magnetic fields rather lower than the anisotropy field of the sample. The spontaneous magnetization is not necessary to be known.

The method has been tested at room temperature on Ba-ferrite powder consisting of single-domain particles. The anisotropy field of 12.3 kOe has been obtained, in a very good agreement with the known value of 12.0 kOe [24].

The method has been applied for estimation of the anisotropy field of melt-spun Nd–Fe–B compound (General Motors' MQ powder). The values obtained are in a reasonable agreement with the data for Nd–Fe–B alloys, found in the literature.

## Appendix

Let us consider a S–W system of randomly orientated single-domain particles whose uniaxial anisotropy decreases with temperature. During ZFC and FC curves measurements, the temperature changes monotonically and so does the magnetic field during initial magnetization curve and demagnetizing curve measurement. Thus, any minor thermomagnetic or hysteresis loops are excluded. The initial magnetization state for both ZFC and IMC measurements is that of randomly orientated magnetic moments, which corresponds to the thermally demagnetized sample. Under these conditions, each point on a given ZFC thermomagnetic curve, i.e., the equilibrium magnetization $M^{\mathrm{ZFC}}(H,T)$ (at a given temperature and magnetic field), corresponds to the state of minimum of the reduced energy (2) for all particles. The minimization (for the same temperature and magnetic field) gives the equilibrium magnetization $M^{\mathrm{IMC}}(T,H)$ of the point corresponding to the same $H$ on the IMC, measured at $T$. (The magnetization changes during ZFC plotting can also be regarded as a result of a decrease of the anisotropy field with temperature).[1] A

[1] This is equivalent to a raise of the reduced magnetic field $H/H_a$ [see (2)], leading to an increase of the reduced magnetization $j$ on the ZFC curve. But the same increase of the reduced field produces the raise of the reduced magnetization on the initial magnetization curve.

numerical solution of the problem is described in more detail in [15] and [16].

The equivalency of the points on FC thermomagnetic curves and DMC curves can be explained in a similar way, as in this case the initial states, corresponding to the magnetic saturation at a given temperature, or cooling down the sample from temperature exceeding $T_c$ in a given magnetic field, are the same. This direct relation between the magnetizations at a fixed $T$ of a couple of ZFC and FC curves, and the IMC and DMD, calculated for this temperature [$M_s(T)$ and $K_u(T)$ are known], is illustrated in Fig. 1. Model ZFC and FC curves for $H = 5$ kOe are plotted in Fig. 1(a) as $M_s(T)$ and $K_u(T)$ dependencies are taken to be those for Ba-ferrite [25]. The IMC and DMC for the same system at $T/T_c = 0.66$ are presented in Fig. 1(b).

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